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Origins of African dust transported over the northeastern tropical Atlantic

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Abstract. On the basis of 3 years of daily measurements of mineral dust concentrations at Sal Island (Cape Verde), this study focuses on the origin of the dust transported over the northeastern tropical Atlantic during the dusty season (October to April). Using backward trajectories, we have identified three different sectors of origin of the transported dust, covering the Sahara (north/west and central/south) and the Sahel regions. Time variations of the dust transport from these different areas can be explained by the seasonal shift of the subtropical high-pressure center. A geochemical approach highlights that the calcium amount and the Si/Al ratio of the transported dust differ according to these source regions, in relation to their respective soil compositions.

Introduction

Mineral dust emitted by arid and semiarid regions of the Earth represent about 45% of the global aerosol emissions [Andreae, 1994]. The Sahara Desert is the most important producer of this soil-derived dust, with annual emissions estimated from 300 to 700 megatons per year [Schütz, 1980; d'Almeida, 1986; Pye, 1987; Marticorena and Bergametti, 1996]. One important characteristic of Saharan dust particles is their ability to be transported over long distances from their source regions. The major part is exported westward and southward [d'Almeida, 1986], over the north tropical and equatorial Atlantic Ocean. For instance, it is well known that the dust can reach the Caribbean Sea after a transport of more than 5000 km over the Atlantic Ocean [Delany et al., 1967; Prospero, 1968; Prospero and Nees, 1977].

As a consequence, many atmospheric and oceanic processes are affected by the transport and deposition of mineral dust. During their atmospheric transport, these particles have the potential for playing an important role in radiative processes and climate both by backscattering the solar radiation and by absorbing the solar and infrared radiation [Intergovernmental Program on Climate Change, 1994; Tegen et al., 1996; Li et al., 1996; Andreae, 1996]. The incorporation of dust particles in cloud droplets also affects the formation of precipitation and its ionic content [Buat-Ménard and Duce, 1986; Loye-Pilot et al., 1986; Losno et al.,

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1991]. The deposition of mineral dust in the ocean is responsible for a large part of the deep-sea sedimentation [Chester et al., 1979; Sarnthein and Koopman, 1980; Rea et al., 1985] and provides nutrients to sea surface waters [Duce et al., 1991; Bergametti et al., 1992].

Being able to identify the dust source regions, their geographical locations, and the temporal variations of their emissions would allow one to provide validation data and to constrain the source schemes used in model simulations. Identifying geochemical tracers associated with the source regions will also contribute to the interpretation of the origin of the dust events recorded at long-term dust-monitoring stations and then to obtaining a climatological view of the source regions. Especially, it is of great interest to determine how areas potentially under strong anthropogenic pressure, like the Sahel, contribute to the present dust transport over the north tropical Atlantic. The identification of geochemical signatures associated with the source regions in relation to meteorological situations is necessary to the interpretation of the paleocirculation from dust sediments. It would allow one to relate past, present, and future changes in the dust content of the tropical Atlantic to changes in both strength and geographical origin of the sources. Complementarily, if changes in dust composition according to their source regions are determinated, the estimation of atmospheric inputs to sea surface water could be improved, especially by providing more accurate estimations for species having a high biological impact, such as iron.

Recently, several studies based on satellite images [N'Doumé, 1993; Legrand et al., 1994] or modeling approach [Marticorena et al., 1997] have investigated at large spatial and temporal scales the main regions producing mineral dust over the west African continent. However, such climatological

studies do not allow establishment of a direct link between these source regions and the main transport directions. A complementary approach consists of identifying the geographical origin of the mineral dust events transported over selected oceanic regions, as done, for example, over the Mediterranean sea [Bergametti et al., 1989a]. In this way the Cape Verde Islands are of particular interest, since they are located within the main transport area of dust over the tropical Atlantic [Schütz, 1980] and close enough to the African coast (about 500 km off Dakar, Senegal) to allow a good identification of the dust source regions over the Sahara desert and Sahel. Up to now, only a few studies have documented the origin of dust transported over the northeastern Atlantic. Two field campaigns were performed in Canary Islands during spring and summer [Coudé-Gaussen et al., 1987; Bergametti et al., 1989b] in order to investigate the source regions of African dust transported over the subtropical Atlantic. These studies strongly suggested that the Moroccan area is one of the main source regions of dust affecting the Canary Islands, but because of the limited duration of these campaigns the representativeness of the identified source regions over larger timescales remains questionable. Concerning the western tropical Atlantic, Prospero et al. [1970] investigated the origin and mineralogical composition of a dust event observed at Barbados in June; on the basis of similarities in composition and air parcel trajectories they concluded that the dust transported during summer and early fall over this region could be derived from the same general area covering Spanish Sahara, Mauritania, Mali and Senegal.

Based on 3 years of continuous measurements of daily concentrations at Sal island (Cape Verde Islands), the present study provides a climatological assessment of the source regions of dust transported over the northeastern tropical Atlantic during the dusty season (October to April). Both geochemical tracers and air mass trajectories are used to assess the origin of dust and the meteorological situations responsible for changes in dust source locations.

Sampling and Analysis

Sal Island (22°57'W, 16°45'N) is the most northeastern island of the Cape Verde archipelago, located 500 km west of the African coast. Beginning in December 1991, aerosol samples have been collected on a daily basis by bulk filtration by using 0.4-µm-pore-size Nuclepore filters, with a flow rate of about 1 m³/h. The sampling system allows the collection of particles smaller than 20 µm in diameter. The sampling site is located at the top of a 25-m-high tower situated at an altitude of 100 m, 5 km from the eastern coastline of the island.

The samples were analyzed by X ray fluorescence spectrometry, following the method described by Losno et al. [1987]. The major elements of soil dust (Si, Al, Fe, Ca, and K) were analyzed, since their relative abundance constitutes useful fingerprints of the aerosol composition according to its origin [Coudé-Gaussen et al., 1987; Bergametti et al., 1989a, 1989b]. Sodium as an indicator of the sea-salt concentrations was used in order to make corrections of the marine components for calcium and potassium. In most cases these corrections were minor, the marine components representing less than 10% of the global Ca and K concentrations during the intense dust events. Analytical precision ranged between 3% and 10%, depending on the element and the concentration level.

Seasonal Aspects of Dust Transport Over the North Tropical Atlantic

Two principal processes of mineral dust transport over the north tropical Atlantic have been identified; in summer, dust transport occurs at high altitudes, within the so-called "Saharan air layer" located above the trade winds inversion, from about 1.5 km to 5-7 km above sea level [Prospero and Carlson, 1972; Carlson and Prospero, 1972]. This transport process allows a long-range transport and controls the seasonal variation of the mineral dust concentrations over the western tropical north Atlantic, where the highest dust concentrations are observed during this period [Prospero and Nees, 1977]. By contrast, because this transport occurs at a high altitude, it does not significantly affect the dust concentrations at ground level in the eastern Atlantic. Indeed, over this region the maximum dust concentrations occur during the winter months (December and January), because of an additional lower layer transport, which takes place in the trade winds layer, below 1.5 to 3 km in altitude [Chiapello et al., 1995].

The daily silicon concentrations (used as a tracer of mineral dust) measured at Sal are reported in Figures 1a, 1b, and 1c for 1992, 1993 and 1994 respectively. The late spring and summer dust transport cases (from May to September), as recorded at ground levels, are infrequent, usually of weak intensity and short duration, compared with those observed in winter. During summer the dust concentrations measured at ground level at Sal are probably poorly representative of the vertically integrated amount of dust in the atmosphere.

Consequently, the study of dust source regions will be limited to the period October to April. During this period, because of the low-altitude transport process over several hundred kilometers from the African coast, an homogenous concentration over the mixing layer can be assumed. Thus the dust concentrations measured at ground level at Sal will be considered representative of the atmospheric dust content. Moreover, only the samples exhibiting silicon concentrations higher than 5 μ g/m³ will be considered (i.e., 199 samples for the 3-year record) and investigated as "dusty days".

Backward Trajectory Approach

In the atmosphere, back-trajectories have proved very useful to analyze either punctual measurements or synoptic variations of a tracer at a specific site [e.g., Miller et al., 1993; Ramonet et al., 1996]. In particular, they have been successfully used for identifying the Saharan source regions of dust collected far from their sources [Prospero et al., 1970; Coudé-Gaussen et al., 1987; Bergametti et al., 1989a, bl. Here we used the TM2z version [Ramonet and Monfray, 1996] of the three-dimensional atmospheric transport model initially developed by Heimann [1995] to calculate threedimensional isosigma back-trajectories with a horizontal resolution of 2.5°x2.5°. The analyzed wind fields from the European Center for Medium-Range Weather Forecasting (ECMWF) are inverted and used as input data to evaluate the dispersion of an initial small parcel emitted at a pinpoint location. The dispersion of this tracer mass in the TM2z model produces a retrodispersion plume growing during 5 days. Twice a day (0000 and 1200 UTC) the position of the center of gravity of the tracer plume is calculated. We also save the distribution of the tracer plume indicating the

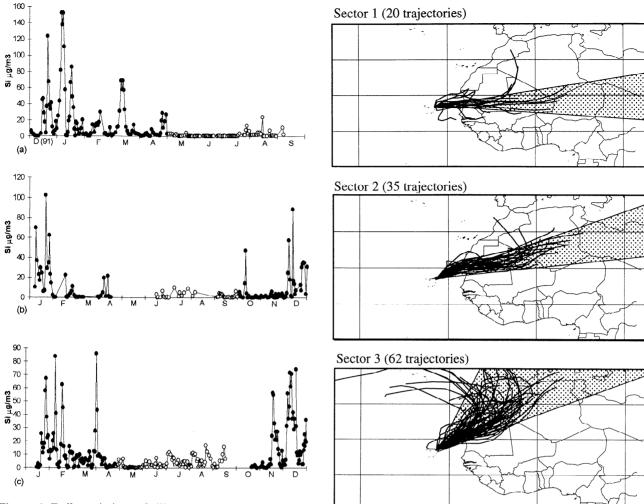


Figure 1. Daily variations of silicon concentrations measured at Sal Island for (a) December 1991 to September 1992, (b) 1993, and (c) 1994. The periods corresponding to the dust source regions study (October to April) are indicated by solid circles.

Figure 2. Sectors of origin of the backward air mass trajectories (approximately 0.4-1.2 km height) associated to the dusty days at Sal Island (1992-1993-1994). Sector 1, Sahel sector, sector 2, south and central Sahara sector, and sector 3, north and west Sahara sector.

qualitative dispersion of air in the area of the trajectory. This method is a powerful tool to analyze atmospheric measurements resulting from complex air mixing upstream at the measurement site. These kinds of situations are not directly resolved by Lagrangian calculations, unless multiple runs are performed with close starting locations. Our trajectories have been successfully compared with other trajectory calculation methods [Ramonet et al., 1996].

For this study we computed trajectories ending above the station in the second layer of the model (approximately 0.4-1.2 km height) according to the low altitude of the dust transport during the period investigated (upper limit between 1.5 and 3 km) [Chiapello et al., 1995]. Also, it must be noted that most of these trajectories when associated with "nondusty samples" present an oceanic origin. A trajectory has been associated with each sample corresponding to a dusty day (generally the trajectory at 0000 hours, since most of the samples are running from about 1200 hours to 1200 hours). Some samples for which the trajectories indicated either strong changes of direction during the 5 days (thus not corresponding to stable situations) or oceanic origin (for most cases at the end of a transport period, when dust concentrations are decreasing)

were discarded. The remaining samples (117) have been classified according to the path and origin of the air masses (as indicated by the trajectories) in three different sectors. Figure 2 illustrates the selected trajectories classification. The three defined sectors can be roughly described as follows: sector 1, the so-called "Sahel sector" (south of the twentieth parallel), including Senegal, south Mauritania, and south Mali; sector 2, the so-called "south and central Sahara sector" (north of twentieth parallel), including Mauritania, north Mali, and south Algeria; and sector 3, the so-called "north and west Sahara sector", including Spanish Sahara, Morocco, and west Algeria.

These sectors are relatively large, and thus some common areas can exist between two sectors (particularly involving the intermediate sector). Nevertheless, the classification of a trajectory within a sector always implies that the major part of its path was made inside this sector. By defining the dust event as a continuous period of dusty days for which the trajectories originate from the same sector, 37 dust events have been classified among the 47 recorded at Sal during the 3 years investigated (i.e., almost 80% of the events).

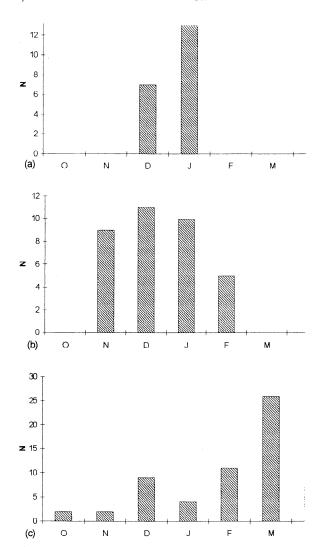


Figure 3. Occurrences of dusty days at Sal Island month by month (from October to April) according to the sectors of origin presented in Figure 2: (a) sector 1, (b) sector 2, and (c) sector 3.

Time Variations of Origin of Transported Dust: Meteorological Situations Implied

From this classification we first examined whether the different origins of the transported dust could correspond to particular periods of the year. According to the three sectors defined above, Figure 3 presents the monthly occurrences of dusty days at Sal (October to April). It clearly appears that dust originating from the Sahel sector corresponds exclusively to events observed at Sal in December and January. This is also the case for the major part (60%) of the dust events originating from the south and central Sahara sector, with frequent events occurring also in November and February. The dust events originating from the north and west Sahara sector are observed all along the dusty season but are dominant in March. Moreover, during October, March, and April, all the dust events observed at Sal originate from this sector. These results strongly suggest a seasonal cycle in the coupled system source/transport of the African dust, which could be connected to the meteorological changes occurring during the year.

An analysis of the meteorological maps of surface pressure [European Meteorological Bulletin, 1992] associated with the observed dust events shows that the transport of dust from these different African areas is related to the location of the subtropical high-pressure centers (the most important being the Azores high). Figures 4a and 4b present typical meteorological situations corresponding to dust transport from the Sahel sector and the north and west Sahara sector respectively. The dust transport from the Sahelian areas is characterized by the high-pressure center located over the Mediterranean Sea and the northern part of Africa, whereas the transport from the northwestern Sahara is observed when the Azores high-pressure center is located off the southwest of Spain, over the Atlantic Ocean. Except for a few unexplained cases this relationship is valid for all the observed dust events and can be considered as a strongly dominant pattern.

Thus the temporal variations of the origin of the dust transport over the northeastern tropical Atlantic appear to be related to the oscillations of the Azores high-pressure center location during the year; in December and January the Azores high is located at its most southerly position and allows the dust transport from sources in Sahel and the southern part of the Sahara. In March and April the seasonal shift of the Azores high toward the northwest has begun, and the transport takes place from the northwestern part of the Sahara. Obviously, these situations reflect average seasonal patterns, and the position of the Azores high can present variations at shorter timescales: this behavior explains, for example, that the dust transport from the north and west Sahara can be observed all along the study period, from October to April.

The remaining question is whether these observed source/transport temporal variations are consistent with the seasonal shifts of the source region locations over Sahara and Sahel. Using Meteosat satellite observations in the thermal infrared channel over the period 1984-1991, N'Doumé [1993] has established a climatology of the dust content over west Africa, which provides information on the spatial and temporal variability of the dust emissions. He observed that the Sahelian region produces dust mainly during December and January, a period during which the Intertropical Front reaches its most southerly position (around 5°N). Goudie and Middleton [1992] also observed maximal frequencies of dust storms in the Sahel region between December and February. From late winter to late spring, N'Doumé noted that the dust source regions move toward the north of the Sahara, in relation to the seasonal shift of the Intertropical Front over Africa. Thus the temporal variations of the origin of the dust observed at Sal seem to reflect the shift of the source region locations over Sahara and Sahel during the year.

Geochemical Study

First, we examined the intensities and frequencies of the dust events at Sal according to their sectors of origin. Figure 5 presents the mean silicon concentrations of events associated with the sectors of origin previously defined. Note that the mean silicon concentration is much greater for dust originating from the Sahel sector (around $67 \, \mu g/m^3$) than from the north and west Sahara sector (around $19 \, \mu g/m^3$), the south and central Sahara sector exhibiting an intermediate value (33 $\, \mu g/m^3$). Concerning frequencies, the observed dust events do not present an homogeneous distribution. Dust events

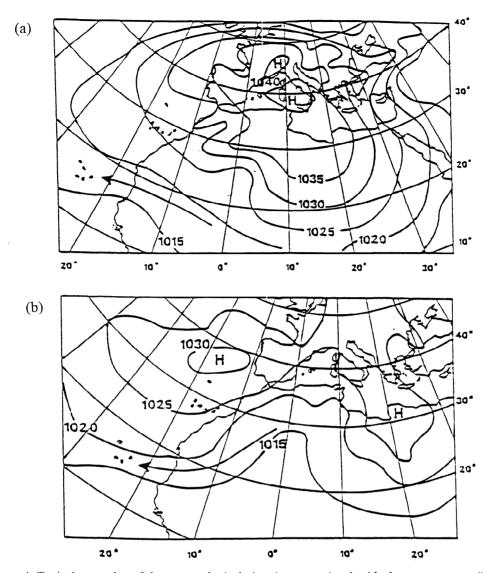


Figure 4. Typical examples of the meteorological situations associated with dust transport at Sal from (a) sector 1 (Sahel), and (b) sector 3 (north and west Sahara). From the *European Meteorological Bulletin* [1992].

originating from north and west Sahara represent more than half the total dust events observed at Sal during the period October-April. The dust events originating from the Sahelian region are few (about 11% of the total of the observed dust events), whereas 32% of the observed dust events at Sal originate from the south and central Sahara. So these results show that if dust events of north and west origin are the more frequent, they are in average less intense in comparison with events originating from south and central Sahara and Sahel sectors. The high intensity of dust events of Sahelian origin is particularly interesting, since it means that the Sahelian region, which may be considered at least in part as an anthropogenic dust source [Tegen and Fung, 1995], contributes significantly to the high dust concentration levels observed during winter over the northeastern tropical Atlantic.

Composition of Transported Dust According to Its Origin

To evaluate whether the chemical composition of the dust observed at Sal differs according to its origin, we computed for each sector the elemental ratios of the major dust elements (Si, Al, Fe, Ca, and K). Table 1 shows that the events originating from Sahel present low Si/Al ratios (average of 2.03) compared with those observed for dust originating from the south and central Sahara (2.21) and the north and west Sahara (2.32). Moreover, these results also point out that dust originating from the north and west Sahara have high calcium amounts (high Ca/Al ratios, low Fe/Ca and K/Ca ratios) in comparison with dust originating from Sahel, whereas dust from the south and central Sahara present intermediate values.

Figure 6 presents the Fe/Ca, K/Ca, Si/Al, and Ca/Al ratio values according to the origin of the dust. One can observe that a clear continuous trend appears from the Sahelian to the north and west Saharan sectors for these ratios. The intermediate values of the ratios associated with the south and central Sahara sector are in agreement with its "middle" geographical position (in latitude) in comparison with the two other sectors.

As a comparison, the Ca/Al and Fe/Ca ratios measured for dust events during a campaign performed in the Canary

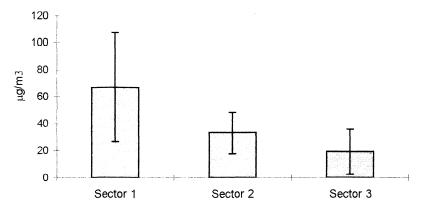


Figure 5. Mean silicon concentrations (μ g/m³) and standard deviations of dust events at Sal Island according to the sectors of origin presented in Figure 2. Variance analysis indicates that one of the means is different at more than 99.9%. From Fisher tests the difference between mean concentrations of sectors 1 and 2 is significant at more than 5%, that between sectors 2 and 3 is significant at more than 1%, and that between sectors 1 and 3 is significant at more than 0.1%.

Islands in July 1985 [Bergametti et al., 1989b] can be used: these values, 1.23 and 1.20 respectively (mean values for silicon concentrations higher than 5 µg/m³), are intermediate to those measured at Sal for dust originating from the north and west Sahara and the south and central Sahara (see Table 1). This finding is consistent with the two identified source regions in Canary Island, covering these two sectors. A good agreement is also observed for dust events originating from north and west Sahara collected in Corsica [Bergametti et al., 1989a], which present mean Si/Al, Ca/Al, and Fe/Ca ratios of 2.71, 0.83 and 0.99 respectively, close to those measured at Sal for dust originating from the same part of the Sahara (sector 3). Globally, these dust ratio values are consistent with the observed geochemical trends at Sal Island (a higher amount of calcium and Si/Al ratios in northern Saharan dust).

Moreover, the study of Chester et al. [1972] has shown, from dust samples collected during oceanic campaigns along the African coast, that the highest kaolinite levels were observed at the lower latitudes. Since kaolinite is the clay mineral exhibiting the lowest Si/Al ratio (around 1) [see Coudé-Gaussen et al., 1987], this mineralogical pattern is consistent with the decrease of Si/Al ratios observed for dust originating from southerly regions (Sahel and south and central Sahara) at Sal. Actually, these observations seem to reflect a constant phenomenon, as indicated by the study of Windom [1975], which points out a significant decrease of the kaolinite content in the North Atlantic deep-sea sediments when latitude increases. Since several authors have shown mineralogical similarities between the composition of African dust and sediments of the North Atlantic [Delany et al., 1967; Windom, 1975; Johnson, 1979], this result can be considered a good indicator of the geographical distribution of African dust composition. In summary, the results obtained at Sal are consistent with the data available regarding dust composition over the northeastern Atlantic, highlighting an evolution of the dust composition as a function of its origin, characterized by an increase of Si/Al ratios and calcium amounts from south to north.

Relation With Dust and Soil Compositions Over the North African Desert

In Figure 6, mean values of the same elemental ratios for dust measured during field campaigns performed in the

Sahara and Sahel have been reported. There is a consistency between elemental ratios measured in dust collected at Sal according to their sector of origin and those obtained for aerosols collected in the corresponding source regions. In particular, the same trend is observed through Sahelian dust being characterized by lower Si/Al ratios (mean, 1.93), compared with central Saharan dust (mean, 2.12) and north Saharan dust (mean, 2.45). In the same way, dust collected in the north Sahara presents higher calcium amounts than dust collected in the south Sahara and Sahelian regions. These data show a consistency between the geochemical signatures of dust collected in source regions and those observed during its transport over the northeastern tropical Atlantic. These results suggest that the dust composition both in source regions and over transport areas could be connected to changes in the soil composition over the north African desert.

We examined the consistency of these geochemical dust fingerprints with the information available concerning the mineralogical characteristics of Saharan and Sahelian soils. In

Table 1. Elemental Ratios of Dust Collected at Sal According to the Sectors of Origin Presented in Figure 2*

Sector 1	Sector 2	Sector 3
2.03	2.21	2.32
0.09	0.07	0.10
0.20	0.36	0.60
0.05	0.07	0.19
2.71	1.43	0.90
0.59	0.32	0.30
0.96	0.62	0.43
0.21	0.10	0.11
	2.03 0.09 0.20 0.05 2.71 0.59	2.03 2.21 0.09 0.07 0.20 0.36 0.05 0.07 2.71 1.43 0.59 0.32 0.96 0.62

^{*} Variance analysis indicates that one of the mean is different at more than 99.9% for the Si/Al, Ca/Al, Fe/Ca and K/Ca ratios. From Fisher tests, the differences between sectors 1 and 2 are significant at more than 0.1% for the Si/Al, Fe/Ca and K/Ca ratios, and 1% for Ca/Al ratio; differences between sectors 2 and 3 and between sectors 1 and 3 are significant at more than 0.1% for all the ratios.

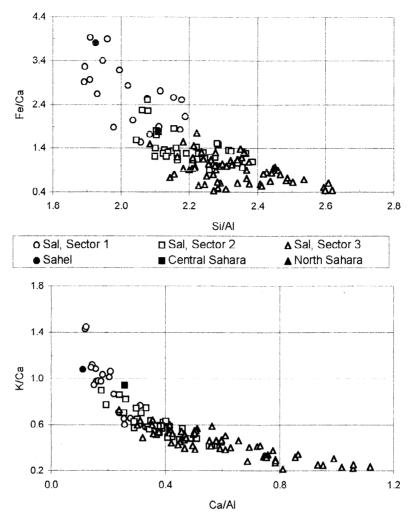


Figure 6. Elemental ratios of dust collected at Sal island according to their sectors of origin (see Figure 2). The solid symbols indicate mean values of elemental ratios for dust measured during field campaigns in the source regions with the same sampling system; Sahel; Niger (M. Sabre and L. Gomes, unpublished data, 1995); central Sahara; Sebbkha Mekkerane (G. Bergametti and P. Raobinarison, unpublished data, 1990); North Sahara; El Abiod, Algeria [Gomes, 1990].

agreement with the Atlantic sediments and dust mineralogical studies of *Windom* [1975] and *Chester et al.* [1972], *Schütz and Sebert* [1987] have shown that kaolinite concentrations are maximal in the soils of the Sahelian belt. This finding can be explained by the influence of lateritic soils, which become more and more frequent in the Sahelian area [*Paquet et al.*, 1984].

The other major geochemical tendency observed at Sal concerns the increase of the amount of calcium in dust originating from the northern Sahara. This is consistent with the mineralogical analysis of Saharan soils performed by Schütz and Sebert [1987] and Gomes [1990], who concluded that a high amount of calcite can be considered as an indicator of dust originating from the north Sahara. Paquet et al. [1984] have also observed high calcite percentages (40-50%) in dust collected in the most northerly part of their track, between the Mediterranean coast and Atlas. These high amounts of calcite may likely be related to the sedimentary depositions of carbonates in North African soils [Schütz and Sebert, 1987; Callot, 1982]. Consequently, these results indicate an influence of the mineralogical characteristics of the source regions soils on the mineral dust composition as measured during its transport over the northeastern tropical Atlantic.

Conclusion

The purpose of this study was to provide information based on a long-time data set allowing us to link the great quantities of dust transported over the northeastern tropical Atlantic to the involved source regions in the North African desert. Over the Cape Verde Islands the maxima of dust concentrations as measured at ground level are observed in winter; this study focused on the dusty season, which can be considered from October to April. Using backward trajectories, we showed that the dust originates from three different sectors covering the Sahara (north/west and central/south) and the Sahel regions. Dust events of north and west Saharan origin are the most frequent, whereas Sahelian dust events appear to be the most intensive; these events occur only during December and January, while in October, March, and April all the dust events observed at Sal originate from the north and west Sahara. These time variations of the dust transport from these different African areas can be explained by the seasonal shift of the subtropical high-pressure center.

A geochemical approach has shown that the calcium amount and the Si/Al ratio of the dust collected at Sal allow us to obtain a clear signature between the different source regions involved. These geochemical fingerprints are consistent with the previous studies of both mineral dust composition and the geographical distribution of sediment compositions over the northeastern Atlantic. Moreover, this study highlights that the differences in dust composition according to its origin, as observed at Sal, can be related to aerosol and soil composition changes over the North African desert.

By providing information on the origins, transport, and concentration levels for the major dust events observed at ground level at Sal island, this work constitutes an excellent reference for the simulations of the dust models. It will allow us to test both their capacity to reproduce the source region locations and the amount and directions of the transported dust. Moreover, these results can be used to refine the biogeochemical cycing of several elements in this region (such as calcium and iron) as a function of the origin of the transported dust.

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